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(54) Title: HERBICIDAL EMULSIFIABLE CONCENTRATE COMPOSITIONS OF DINITROANILINE AND OXYACETAMIDE HERBICIDES

(57) Abstract: The present invention provides herbicidal emulsifiable concentrate compositions of dinitroaniline herbicides and oxyacetamide herbicides. The compositions utilize phosphoric acid as a stabilizing agent to prevent degradation of the oxyacetamide herbicide component during storage.



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HERBICIDAL EMULSIFIABLE CONCENTRATE COMPOSITIONS OF
DINITROANILINE AND OXYACETAMIDE HERBICIDES

BACKGROUND OF THE INVENTION

Dinitroaniline herbicides such as pendimethalin and trifluralin, and oxyacetamide herbicides such as fluthiamid are known in the art. Combination treatments using these types of herbicides are
5 disclosed in U.S. Patents 4,968,342 and 5,759,955; and in German Application No. 19720367. However, emulsifiable concentrate compositions comprising dinitroaniline and oxyacetamide herbicides are not known in the art.

10 U.S. Patent 3,898,075 relates to stabilized liquid compositions containing *m*-biscarbamates and organic acids. U.S. Patent 2,954,396 describes a method for stabilizing carbamate esters with certain hydrolysis inhibitors. However, neither U.S. Patent 3,898,075 nor
15 U.S. Patent 2,954,396 describe any method for the stabilization of oxyacetamide herbicides.

It is therefore an object of the present invention to provide stable emulsifiable concentrate compositions of dinitroaniline herbicides and oxyacetamide
20 herbicides.

Other objects and advantages of the present invention will be apparent to those skilled in the art from the description below and the appended claims.

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SUMMARY OF THE INVENTION

The present invention relates to a herbicidal emulsifiable concentrate composition comprising, on a weight to weight basis, about 15% to 40% of a
5 dinitroaniline herbicide, about 1% to 10% of an oxyacetamide herbicide, about 0.01% to 1% phosphoric acid, up to about 30% of an emulsifier or mixture of emulsifiers, up to about 1% of an antifoaming agent, and an organic solvent or mixture of organic solvents.
10

DETAILED DESCRIPTION OF THE INVENTION

The herbicidal emulsifiable concentrate compositions of this invention comprise, on a weight to
15 weight basis, about 15% to 40% of a dinitroaniline herbicide, about 1% to 10% of an oxyacetamide herbicide, about 0.01% to 1% phosphoric acid, up to about 30% of an emulsifier or mixture of emulsifiers, up to about 1% of an antifoaming agent, and an organic
20 solvent or mixture of organic solvents.

Preferred herbicidal emulsifiable concentrate compositions of the present invention are those comprising, on a weight to weight basis, about 15% to 40% of a dinitroaniline herbicide, about 1% to 10% of
25 an oxyacetamide herbicide, about 0.01% to 1% phosphoric acid, about 2% to 10% of an alkylarylsulfonate, about 0.5% to 5% of a polyalkylene glycol ether, about 1% to 5% of an ethoxylated fatty alcohol, up to about 1% of an antifoaming agent, and an aromatic hydrocarbon
30 solvent having a distillation range of about 135°C to 305°C.

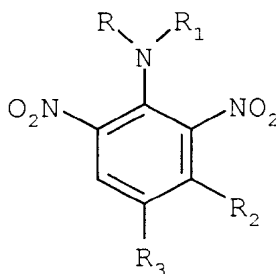
More preferred emulsifiable concentrate compositions of this invention are those comprising, on a weight to weight basis, about 25% to 35%
35 pendimethalin, about 4% to 8% fluthiamid, about 0.05%

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to 0.2% phosphoric acid, about 2% to 6% of a dodecylbenzenesulfonate, about 1% to 3% of an alkyl capped ethylene oxide/propylene oxide block copolymer, about 1% to 5% of an ethoxylated C₁₀-C₁₆ fatty alcohol
5 having about 8 to 12 moles of ethylene oxide per mole, up to about 1% of an antifoaming agent, and an aromatic hydrocarbon solvent having a distillation range of about 135° to 305°C.

Phosphoric acid is an especially important element
10 of the present compositions because it has been found that oxyacetamide herbicides are more stable in the phosphoric acid containing emulsifiable concentrate compositions of the present invention than in
15 corresponding emulsifiable concentrate compositions which do not contain phosphoric acid. In a preferred embodiment of the present invention, the ratio of the oxyacetamide herbicide to the phosphoric acid, on a weight basis, is preferably about 30:1 to 100:1, and more preferably, is about 40:1 to 70:1.

20 Dinitroaniline herbicides suitable for use in the compositions of this invention have the structural formula I



(I)

25 wherein

R is hydrogen, C₂-C₄alkyl or chloroethyl;

R₁ is C₂-C₅alkyl, chloroethyl, 2-methallyl or cyclopropylmethyl;

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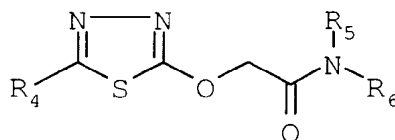
R₂ is hydrogen, methyl or amino; and

R₃ is trifluoromethyl, C₁-C₃alkyl, SO₂NH₂ or SO₂CH₃.

Preferred dinitroaniline herbicides for use in this invention include pendimethalin and trifluralin
5 with pendimethalin being more preferred.

Oxyacetamide herbicides suitable for use in the compositions of the present invention are those described in U.S. Patent Nos. 4,408,055; 4,509,971; 4,540,430; 4,549,899; 4,585,471; 4,645,525; 4,756,741;
10 4,784,682; 4,788,291; 4,833,243; 4,968,342; 4,988,380; 5,090,991 and 5,234,896; EP Application Nos. 300344-A and 500934-A1; and WO 97/08160.

Preferred oxyacetamide herbicides for use in the compositions of this invention have the structural
15 formula II



(II)

wherein

20 R₄ is hydrogen, F, Cl, Br, C₁-C₄alkyl, C₁-C₄haloalkyl, C₁-C₄alkoxy, C₁-C₄haloalkoxy, C₁-C₄alkylsulfonyl or C₁-C₄haloalkylsulfonyl;

R₅ is hydrogen or C₁-C₆alkyl; and

R₆ is phenyl optionally substituted with any
25 combination

of from one to three F, Cl, Br, CN, NO₂, C₁-C₄alkyl,

C₁-C₄haloalkyl, C₁-C₄alkoxy or C₁-C₄haloalkoxy groups.

30 More preferred oxyacetamide herbicides for use in the emulsifiable concentrate compositions of this invention are those wherein

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R₄ is C₁-C₄haloalkyl;

R₅ is C₁-C₄alkyl; and

R₆ is phenyl optionally substituted with any combination

5 of one or two F, Cl or C₁-C₄alkyl groups.

Most preferred oxyacetamide herbicides are those of formula II wherein

R₄ is CF₃, CHF₂ or CFCl₂;

R₅ is isopropyl; and

10 R₆ is selected from the group consisting of phenyl, 3-chlorophenyl, 4-chlorophenyl, 2-fluorophenyl, 4-fluorophenyl, 2-methylphenyl, 3-methylphenyl, 4-methylphenyl and 3,4-dimethylphenyl.

An oxyacetamide herbicide which is especially
15 suitable for use in the compositions of this invention is 4'-fluoro-N-isopropyl-2-{[5-(trifluoromethyl)-1,3,4-thiadiazol-2-yl]oxy}acetanilide (fluthiamid).

Emulsifiers suitable for use in the compositions of the present invention include, but are not limited
20 to, alkylarylsulfonates including C₈-C₁₈alkylbenzenesulfonates such as calcium dodecylbenzenesulfonate; polyalkylene glycol ethers including alkyl capped ethylene oxide/propylene oxide block copolymers such as butyl capped ethylene
25 oxide/propylene oxide block copolymers; and ethoxylated fatty alcohols including ethoxylated C₁₀-C₁₆fatty alcohols having about 8 to 12 moles of ethylene oxide per mole; and mixtures thereof. Preferred emulsifiers include the alkali metal or
30 alkaline earth metal salts of dodecylbenzene sulfonic acid, such as calcium dodecylbenzenesulfonate including SOPROPHOR[®] 70 a 60% calcium dodecylbenzene sulfonate solution in isobutanol commercially available from Rhodia, Milan, Italy; butyl capped ethylene
35 oxide/propylene oxide block copolymers including

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WITCONOL[®] NS 500K a butyl capped ethylene oxide/propylene oxide block copolymer commercially available from Witco SA, Saint-Pierre Les Elbeuf, France; and ethoxylated C₁₀-C₁₆ fatty alcohols having
5 about 8 to 12 moles of ethylene oxide per mole including RHODASURF[®] 870 a C₁₃ ethoxylated fatty alcohol containing 10 moles of ethylene oxide per mole commercially available from Rhodia, Milan, Italy; and mixtures thereof.

10 Organic solvents suitable for use in the compositions of this invention include aromatic hydrocarbon solvents such as toluene, xylenes, polynuclear aromatic hydrocarbons such as naphthalenes and alkylnaphthalenes and mixtures thereof, many of
15 which are available from the fractionation of crude oil and in general have distillation ranges in the temperature range of about 135°C to 305°C, with those having a distillation range of from about 183°C to 290°C being most preferred. These solvents are
20 commercially available under a variety of tradenames, e.g. SOLVESSO[®] 200 and AROMATIC[®] 200 both commercially available from Exxon, Fareham, Hants, United Kingdom.

Antifoaming agents suitable for use in the compositions of the present invention include
25 conventional antifoaming agents, with silicone based antifoaming agents such as those sold under the SILCOLAPSE[®] tradename commercially available from Rhodia, Lyon, France being preferred. In a preferred embodiment of this invention, an antifoaming agent is
30 used at a level sufficient to prevent undesirable foaming during the preparation of tank mixes using the emulsion concentrates of the present invention. Typically, less than 1% by weight of a defoamer is sufficient, with amounts of about 0.01 to about 0.1% by
35 weight being preferred.

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The herbicidal emulsifiable concentrate compositions of the present invention may be prepared by admixing all of the ingredients in the organic solvent. In a preferred embodiment of this invention,
5 the compositions are prepared by:

(a) admixing an alkylarylsulfonate, an ethoxylated fatty alcohol, an antifoaming agent and an organic solvent to obtain a first homogeneous mixture;

(b) adding a molten dinitroaniline and a molten
10 polyalkylene glycol ether to the first homogenous mixture of step (a) with stirring to obtain a second homogenous mixture;

(c) adding an oxyacetamide herbicide to the second homogenous mixture of step (b) with stirring to obtain
15 a third homogenous mixture; and

(d) adding phosphoric acid to the third homogenous mixture of step (c) with stirring.

The emulsifiable concentrate compositions of this invention are diluted with water and applied as dilute,
20 aqueous emulsions to the locus where weed control is desired. Typical dilution rates are in the range of about 1 L of concentrate per 400 L of water to 4 L of concentrate per 100 L of water. While the compositions of this invention are effective for controlling weeds
25 when employed alone, they may also be used in conjunction with or in combination with other biological chemicals, including other herbicides.

In order to facilitate a further understanding of the invention, the following examples are presented
30 primarily for the purpose of illustrating more specific details thereof. The scope of the invention should not be deemed limited by the examples, but encompasses the entire subject matter defined in the claims.

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EXAMPLE 1Preparation of emulsifiable concentrate compositions

5 A mixture of a 60% calcium dodecylbenzene sulfonate solution in isobutanol commercially available as SOPROPHOR[®] 70 from Rhodia, Milan, Italy (53.0g, 5.00% on a weight basis), a C₁₃ ethoxylated fatty alcohol containing 10 moles of ethylene oxide per mole
10 commercially available as RHODASURF[®] 870 from Rhodia, Milan, Italy (32.0g, 3.00% on a weight basis), a silicone antifoam commercially available as SILCOLAPSE[®] 431 from Rhodia, Lyon, France (0.1g, 0.01% on a weight basis), and an aromatic hydrocarbon mixture (C₁₀-
15 C₁₃)aromatics having a distillation range of about 226-279°C commercially available as SOLVESSO[®] 200 from Exxon, Fareham, Hants, United Kingdom (557.0g, 52.29% on a weight basis) is stirred until homogeneous. Molten pendimethalin (333.3g, 90% real, 31.33% on a
20 weight basis) and a molten butyl capped ethylene oxide/propylene oxide block copolymer commercially available as WITCONOL[®] NS 500K from Witco SA, Saint-Pierre Les Elbeuf, France (21.0g, 2.00% on a weight basis) are added to the mixture with stirring. Solid
25 fluthiamid (66.7g, 90% real, 6.27% on a weight basis) is then added to the mixture and the mixture is stirred until all of the solids dissolve. An 85% phosphoric acid solution (1.0g, 0.10% on a weight basis) is added to the mixture, with stirring until homogeneous, to
30 obtain the emulsifiable concentrate composition identified as composition number 1 in Table I.

 Using essentially the same procedure, the emulsifiable concentrate compositions identified as composition numbers 2 to 14 in Table I are obtained.

TABLE I
Emulsifiable Concentrate Compositions

Comp. No.	Ingredient-%w/w									
	<u>Pendimethalin</u>	<u>Fluthiamid</u>	<u>85% Phosphoric Acid Solution</u>	<u>SOPROPHOR 70</u>	<u>RHODASURF 870</u>	<u>WITCONOL NS 500K</u>	<u>SILCOLAPSE 431</u>	<u>SOLVESCO 200</u>		
1	31.33 ¹	6.27 ¹	0.10	5.00	3.00	2.00	0.01	52.29		
2	28.20 ²	5.64 ²	0.10	5.00	3.00	2.00	0.01	56.05		
3	32.00 ¹	6.40 ¹	0.05	3.84	2.60	1.06	0.01	54.04		
4	32.00 ¹	6.40 ¹	0.10	3.84	2.60	1.06	0.01	53.99		
5	32.00 ¹	6.40 ¹	0.15	3.84	2.60	1.06	0.01	53.94		
6	32.00 ¹	6.40 ¹	0.20	3.84	2.60	1.06	0.01	53.89		
7	32.00 ¹	6.40 ¹	0.25	3.84	2.60	1.06	0.01	53.84		
8	32.00 ¹	6.40 ¹	0.50	3.84	2.60	1.06	0.01	53.59		
9	28.80 ²	5.76 ²	0.05	3.84	2.60	1.06	0.01	57.88		
10	28.80 ²	5.76 ²	0.10	3.84	2.60	1.06	0.01	57.83		
11	28.80 ²	5.76 ²	0.15	3.84	2.60	1.06	0.01	57.78		
12	28.80 ²	5.76 ²	0.20	3.84	2.60	1.06	0.01	57.73		
13	28.80 ²	5.76 ²	0.25	3.84	2.60	1.06	0.01	57.68		
14	28.80 ²	5.76 ²	0.50	3.84	2.60	1.06	0.01	57.43		

¹90% real²100% real

SUBSTITUTE SHEET (RULE 26)

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EXAMPLE 2**Evaluation of storage stability of test compositions**

Samples of test compositions identified in Table I and a comparative composition identified below are placed in glass bottles. The bottles are capped and placed in a temperature controlled storage incubator. After completing the required storage period, the samples are removed and assayed for pendimethalin and fluthiamid content. The active ingredient content is expressed as a percentage relative to the active ingredient content prior to storage (% Initial Content). The results are summarized in Table II.

As can be seen from the data in Table II, fluthiamid is more stable in the emulsifiable concentrate compositions of the present invention which contain phosphoric acid than the comparative emulsifiable concentrate composition which does not contain phosphoric acid.

COMPARATIVE COMPOSITION¹

<u>Ingredient</u>	<u>%w/w</u>
Pendimethalin (90% real)	32.00
Fluthiamid (90% real)	6.40
SOPROPHOR [®] 70	3.84
RHODASURF [®] 870	2.60
WITCONOL [®] NS 500K	1.06
SILCOLAPSE [®] 431	0.01
SOLVESSO [®] 200	54.09

¹Prepared according to the procedure described in Example 1 except that the 85% phosphoric acid solution is not added.

Table II
Evaluation of Storage Stability

Composition Number	Storage Temperature (°C)	Storage Period (weeks)	85% Phosphoric Acid Solution (% w/w)	% Initial Content	
				Pendimethalin	Fluthiamid
Comparative Composition	54	2	0	100	95
3	54	2	0.05	100	100
4	54	2	0.10	100	99
5	54	2	0.15	101	99
6	54	2	0.20	100	98
6	37	12	0.20	99	98
7	54	2	0.25	103	103
8	54	2	0.50	102	102
Comparative Composition	37	12	0	101	94
3	37	12	0.05	100	99
4	37	12	0.10	99	101
5	37	12	0.15	99	99
6	37	12	0.20	99	98

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WHAT IS CLAIMED IS:

1. A herbicidal emulsifiable concentrate composition which comprises, on a weight to weight basis, about 15% to 40% of a dinitroaniline herbicide, about 1% to 10% of an oxyacetamide herbicide, about 0.01% to 1% phosphoric acid, up to about 30% of an emulsifier or mixture of emulsifiers, up to about 1% of an antifoaming agent, and an organic solvent or mixture of organic solvents.

2. The composition according to Claim 1 wherein the emulsifier or mixture of emulsifiers comprises about 2% to 10% of an alkylarylsulfonate, about 0.5% to 5% of a polyalkylene glycol ether and about 1% to 5% of an ethoxylated fatty alcohol, and the organic solvent or mixture of organic solvents is an aromatic hydrocarbon solvent having a distillation range of about 135°C to 305°C.

3. The composition according to Claim 1 which comprises, on a weight to weight basis, about 25% to 35% pendimethalin, about 4% to 8% fluthiamid, about 0.05% to 0.2% phosphoric acid, about 2% to 6% of a dodecylbenzenesulfonate, about 1% to 3% of an alkyl capped ethylene oxide/propylene oxide block copolymer, about 1% to 5% of an ethoxylated C₁₀-C₁₆ fatty alcohol having about 8 to 12 moles of ethylene oxide per mole, up to about 1% of an antifoaming agent, and an aromatic

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hydrocarbon solvent having a distillation range of about 135°C to 305°C.

4. The composition according to Claim 1 wherein the ratio of the oxyacetamide herbicide to the phosphoric acid on a weight basis is about 30:1 to 100:1.

5. The composition according to Claim 4 wherein the ratio is about 40:1 to 70:1.

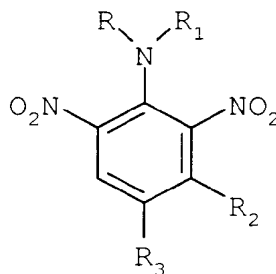
6. The composition according to Claim 1 wherein the emulsifier is selected from the group consisting of an alkylarylsulfonate, a polyalkylene glycol ether, and an ethoxylated fatty alcohol, and mixtures thereof.

7. The composition according to Claim 6 wherein the alkylarylsulfonate is a C₈-C₁₈alkylbenzenesulfonate; the polyalkylene glycol ether is an alkyl capped ethylene oxide/propylene oxide block copolymer, and the ethoxylated fatty alcohol is an ethoxylated C₁₀-C₁₆fatty alcohol having about 8 to 12 moles of ethylene oxide per mole.

8. The composition according to Claim 1 wherein the antifoaming agent is present.

9. The composition according to Claim 1 wherein the dinitroaniline herbicide has the structural formula I

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(I)

wherein

R is hydrogen, C₂-C₄alkyl or chloroethyl;

R₁ is C₂-C₅alkyl, chloroethyl, 2-methallyl or cyclopropylmethyl;

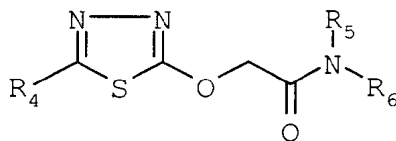
R₂ is hydrogen, methyl or amino; and

R₃ is trifluoromethyl, C₁-C₃alkyl, SO₂NH₂ or SO₂CH₃.

10. The composition according to Claim 1 wherein the dinitroaniline herbicide is selected from the group consisting of pendimethalin and trifluralin.

11. The composition according to Claim 1 wherein the dinitroaniline herbicide is pendimethalin.

12. The composition according to Claim 1 wherein the oxyacetamide herbicide has the structural formula II



(II)

wherein

R₄ is hydrogen, F, Cl, Br, C₁-C₄alkyl, C₁-C₄haloalkyl, C₁-C₄alkoxy, C₁-C₄haloalkoxy, C₁-C₄alkylsulfonyl or

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C₁-C₄haloalkylsulfonyl;
R₅ is hydrogen or C₁-C₆alkyl; and
R₆ is phenyl optionally substituted with any combination
of from one to three F, Cl, Br, CN, NO₂, C₁-C₄alkyl,
C₁-C₄haloalkyl, C₁-C₄alkoxy or C₁-C₄haloalkoxy groups.

13. The composition according to Claim 12 wherein
R₄ is C₁-C₄haloalkyl;
R₅ is C₁-C₄alkyl; and
R₆ is phenyl optionally substituted with any combination
of one or two F, Cl or C₁-C₄alkyl groups.

14. The composition according to Claim 13 wherein
R₄ is CF₃, CHF₂ or CFCl₂;
R₅ is isopropyl; and
R₆ is selected from the group consisting of phenyl,
3-chlorophenyl, 4-chlorophenyl, 2-fluorophenyl,
4-fluorophenyl, 2-methylphenyl, 3-methylphenyl,
4-methylphenyl and 3,4-dimethylphenyl.

15. The composition according to Claim 1 wherein
the oxyacetamide herbicide is fluthiamid.

16. The composition according to Claim 1 wherein
the organic solvent is an aromatic hydrocarbon solvent
having a distillation range of about 135°C to 305°C.

17. The composition according to Claim 16 wherein
the distillation range is about 183°C to 290°C.

18. An aqueous tank mixture which comprises water
and a composition as described in Claim 1.

INTERNATIONAL SEARCH REPORT

Inter: nal Application No
PCT/US 00/16947

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 A01N43/78 A01N43/82 A01N33/18 A01N25/22 //(A01N33/18,
43:82,43:78)

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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A	WO 94 02014 A (BAYER AG ; SANTEL HANS JOACHIM (DE); FEUCHT DIETER (DE)) 3 February 1994 (1994-02-03) cited in the application page 4, line 13-16 page 10, line 25 -page 11, line 11 ----	1-18
A	EP 0 496 989 A (AMERICAN CYANAMID CO) 5 August 1992 (1992-08-05) page 2, line 14-17 -----	1-18

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

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